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Groundwater arsenic in Chandrapur District, Central India.

Arsénico en aguas subterráneas en el distrito de Chandrapur, India central.

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ABSTRACT

To ascertain the groundwater arsenic concentration from the Chandrapur district of Maharashtra State of Central India grab water sampling was carried out in post-monsoon season from 36 sampling locations comprised of 34 from hand pump and two from dug well. Groundwater samples were analysed for general parameters by adopting standard methods as described in APHA (2017). Furthermore, groundwater arsenic concentration was estimated by using the acid digestion method and by using ICP-OES. The results revealed groundwater arsenic concentration presence in all the sampling locations. The groundwater arsenic concentration was in the range of 0.015-0.041 mg/L with an average concentration of 0.031 mg/L. All the samples had groundwater arsenic concentrations above the acceptable limit of the Indian Standard for arsenic (< 0.01 mg/L), although they were within the permissible limit (< 0.05 mg/L). Maximum (n = 23, 63.88%) groundwater samples had arsenic concentration between 0.03-0.05 mg/L followed by 13 (36.11%) in 0.01-0.03 mg/L. The groundwater arsenic concentration in different aquifers revealed the hand pump sample had a maximum arsenic concentration (0.041 mg/L). Pearson's Correlation Coefficient indicated groundwater arsenic is positively correlated with chloride (0.13) and iron (0.16) and negatively correlated with fluoride (-0.21) and pH (-0.19). The groundwater arsenic concentration, electrical conductivity, and pH relationship revealed maximum groundwater arsenic concentration at <1000 mmhos/cm and >1000 mmhos/cm conductivity is obtained in acidic pH; whereas, minimum in near neutral and alkaline pH. The behaviour of groundwater arsenic below ground level revealed the maximum average concentration of 0.0339 mg/L at 35 feet below ground level (bgl) followed by 0.0328 mg/L at 300 feet bgl and minimum (0.0299 mg/L) at 190 feet bgl. Deep wells (101-150 feet bgl) had maximum average groundwater arsenic concentration (0.0326 mg/L) followed by shallow wells (<100 feet bgl) 0.0319 mg/L. From the results, it can be concluded that the presence of arsenic in the groundwater of study area and its origin can be geogenic in nature. The groundwater arsenic concentration was above the permissible limit and thus may pose a health risk to inhabitants who are drinking groundwater. A long-term study from the study area will shed light on spatio-temporal behaviour of groundwater arsenic. A local policy from the government is required to mitigate adverse impacts on the inhabitants. An alternative source of potable water should be made available to the inhabitants thus reducing chronic daily intake and hazards associated with it. A low-cost, environmentally friendly, easy-to-adopt, and understandable arsenic removal methodology will be a boon to the local people.

Keywords: Arsenic, Chandrapur, Groundwater quality, Heavy metal, India, Trace metal.

RESUMEN

Para determinar la concentración de arsénico en el agua subterránea del distrito de Chandrapur del estado de Maharashtra en la India central, se llevó a cabo un muestreo de agua potable en la temporada posterior al monzón en 36 lugares de muestreo, 34 de bombas manuales y dos de pozos excavados. Se analizaron muestras de agua subterránea para determinar parámetros generales mediante la adopción de métodos estándar como se describe en APHA (2017). Además, la concentración de arsénico en el agua subterránea se estimó mediante el método de digestión ácida y mediante el uso de ICP-OES. Los resultados revelaron la presencia de concentración de arsénico en el agua subterránea en todos los lugares de muestreo. La concentración de arsénico en el agua subterránea estuvo en el rango de 0,015 a 0,041 mg/L con una concentración promedio de 0,031 mg/L. Todas las muestras tenían concentraciones de arsénico en aguas subterráneas superiores al límite aceptable del estándar indio para arsénico (< 0,01 mg/L), aunque estaban dentro del límite permisible (< 0,05 mg/L). Las muestras máximas (n = 23, 63,88%) de agua subterránea tenían una concentración de arsénico entre 0,03-0,05 mg/L seguida de 13 (36,11%) entre 0,01-0,03 mg/L. La concentración de arsénico en el agua subterránea en diferentes acuíferos reveló que la muestra de la bomba manual tenía una concentración máxima de arsénico (0,041 mg/L). El coeficiente de correlación de Pearson indicó que el arsénico del agua subterránea se correlaciona positivamente con el cloruro (0,13) y el hierro (0,16) y negativamente con el fluoruro (-0,21) y el pH (-0,19). La concentración de arsénico del agua subterránea, la conductividad eléctrica y la relación de pH revelaron que la concentración máxima de arsénico en el agua subterránea a <1000 mmhos/cm y la conductividad >1000 mmhos/cm se obtiene en pH ácido; mientras que mínimo en pH casi neutro y alcalino. El comportamiento del arsénico en las aguas subterráneas bajo el nivel del suelo reveló una concentración promedio máxima de 0.0339 mg/L a 35 pies bajo el nivel del suelo (bgl), seguida de 0.0328 mg/L a 300 pies bgl y una mínima (0.0299 mg/L) a 190 pies bgl. Los pozos profundos (101-150 pies bgl) tuvieron una concentración promedio máxima de arsénico en el agua subterránea (0.0326 mg/L), seguidos por los pozos poco profundos (<100 pies bgl) 0.0319 mg/L. De los resultados se puede concluir que la presencia de arsénico en las aguas subterráneas del área de estudio y su origen puede ser de naturaleza geogénica. La concentración de arsénico en las aguas subterráneas estaba por encima del límite permitido y, por tanto, puede suponer un riesgo para la salud de los habitantes que beben aguas subterráneas. Un estudio a largo plazo en el área de estudio arrojará luz sobre el comportamiento espacio-temporal del arsénico en las aguas subterráneas. Se requiere una política local por parte del gobierno para mitigar los impactos adversos sobre los habitantes. Se debe poner a disposición de los habitantes una fuente alternativa de agua potable, reduciendo así el consumo diario crónico y los peligros asociados a ella. Una metodología de eliminación de arsénico de bajo costo, respetuosa con el medio ambiente, fácil de adoptar y comprensible será de gran ayuda para la población local.

Palabras clave: Arsénico, Chandrapur, Calidad de las aguas subterráneas, Metales pesados, India, Trazametales.

INTRODUCTION

The Earths crust (10-70 kilometers) is physically and chemically the most varied geopshere with lowest temperature, thickness, and mass. The crust's chemical composition, include 93 elements, however 8 of them represent as much as 99.5% of its mass. Minerals are not evenally distributed in the Earths crust. The Earth's upper crust contains 1.5-2 mg/kg arsenic, coal between 0.5 and 93 mg/kg with a mean value of 17.7 mg/kg, and

brown coal up to 1500 mg/kg (Finkelman *et al*., 1999). Sulphide deposits sometimes contain ≥60 mg/kg arsenic. Arsenopyrite (FeAsS) is the most abundant ore of arsenic, others include arsenolite, As_2O_3; mimetite, $Pb_5Cl(AsO_4)_3$; olivenite, Cu_2OHAsO_4; cobaltite, CoAsS, proustite, Ag_3AsS_3, lignite ($FeAs_2$), orpigment (As_2S_3) and realgar (AsS). Arsenic also occurs in metallic form (Stoeppler, 2004).

Arsenic concentrations in unpolluted fresh waters, mainly as As^V, commonly range from 1 to 10 mg/L (Mandal and Suzuki, 2002). The average from 100 municipal water supplies in the USA was <10 mg/L, while that from 6000 samples from waterworks studied between 1987 and 1990 in Germany was 0.65 mg/L (Iffland, 1994). In areas of sulphide mineralization and mining, and also in the vicinity of pesticide-producing plants, arsenic contents in drinking water may rise to 0.1-5 mg/L (Mandal and Suzuki, 2002), with extremely high values of up to 58 mg/L found in Calcutta, India (Chatterjee and Chakraborti, 1993).

Mineral waters may contain up to 50 times, and hot springs up to 300 times, more arsenic than normal background levels (Savory and Wills, 1984). Drinking water supplies and groundwater in some regions in South America, Asian countries, USA and Europe contain arsenic levels up to 3.6 mg/L or even higher (Concha *et al*., 1998, Vahter 1999, Ng *et al*., 2003, Tareq *et al*., 2003, Yang *et al*., 2003,). Thus, several millions of people worldwide are at risk of consuming arsenic-contaminated water from natural sources.

Severe poisoning can arise from the ingestion of as little as 100 mg of arsenic trioxide; chronic effects may result from the accumulation of arsenic compounds in the body at low intake levels. Carcinogenic properties also have been imputed to arsenic compounds. The toxicity of arsenic depends on its chemical form. Arsenite is many times more toxic than arsenate. For the protection of aquatic life, the average concentration of As^{3+} in water should not exceed 72 µg/L and the maximum should not exceed 140 µg/L.

Arsenic is estimated to affect more than 150 million people worldwide with its increasingly elevated concentrations in drinking water (Stroud *et al*., 2011). The major arsenicosis-affected areas have been reported in large deltas and/or along major river basins across the world (Fendorf *et al*., 2010) such as in Paraiba do Sul delta, Brazil (Mirlean *et al*., 2014), Bengal delta, India (Mukherjee *et al*., 2006, Chakraborti *et al*., 2010, Shukla *et al*., 2010), Mekong delta, Cambodia (Sthiannopkao *et al*., 2008), Danube river basin, Hungry (Nriagu *et al*., 2007), Hetao river basin, Mongolia (Khan and Ho, 2011), Duero Cenozoic Basin, Spain (Gomez *et al*., 2006), Zenne river basin, Belgium (Nriagu *et al*., 2007), and Tulare Lake, USA (Cutler *et al*., 2013). The transfer of arsenic to the food chain will ultimately remain a long-term risk to human and ecological systems (Tuli *et al*., 2010). Since water is the principal route through which arsenic enters the human body (Chen *et al*., 2009), an understanding of the processes of arsenic contamination in groundwater, associated health risks, and mitigation of arsenic problems is required.

Table 1 presents the details about groundwater arsenic in different countries. From the table, it can be observed that groundwater arsenic is dominant throughout the world from Argentina to Vietnam. Moreover, the groundwater arsenic concentration present is manyfold higher than the permissible limit of the World Health Organisation (10 µg/L). The groundwater arsenic concentration and the population at risk due to ingestion of this groundwater are presented in Table 2. The details presented in the table reveal groundwater arsenic is present in the aquatic environment since 1950 (Taiwan). The maximum groundwater arsenic concentration was in the order of USA (48000) > China (8000) > Thailand (5000) which indicates its concentration is manyfold higher than

the prescribed permissible limit of World Health Organisation (10 µg/L). This higher groundwater arsenic concentration makes the local inhabitants at higher risk. The population of different countries at risk due to ingestion of groundwater arsenic is in the order of Bangladesh (50, 000 000) > Argentina (2000 000) > India (1000 000).

Table 1: Groundwater arsenic in different countries

Country	Region	Concentration	Permissible limit	References
Argentina	Chaco-Pampean (South-western)	13-621 µg/L	10 µg/L (WHO)	Mariño *et al*., (2020)
Bangladesh	South-east part of Bangladesh	>200 µg/L	50 µg/L (Bangladesh standard)	Kinniburgh *et al*., (2003)
China	Tumochuan Plain	200.3 µg/L (Unconfined) 162.3 µg/L (Unconfined)	10 µg/L (USEPA)	Dong *et al*., (2022)
India	West Bengal	10.1-213 µg/L (≥10 µg/L) <3.0-9.7 µg/L (<10 µg/L)	10 µg/L (Indian standard)	De *et al*., (2022)
Iran	Kerman	4-278 µg/L	10 µg/L (USEPA)	Rahnamarad *et al*., (2020)
Pakistan	Bahawalnagar	BDL-31.5 µg/L	10 µg/L (USEPA)	Shahid *et al*., (2023)
Turkey	Suhut Plain	2.65-36.50 µg/L	10 µg/L (WHO)	Varol *et al*., (2021)
Uruguay	Uruguay	1.72-120.48 µg/L	10 µg/L (WHO)	Machado *et al*., (2019)
USA	Michigan	>10 µg/L – 220 µg/L	10 µg/L (USEPA)	Kolker *et al.*, (2003)
USA	New Mexico, Middle Rio Grande Basin	<1-600 µg/L	10 µg/L (USEPA)	Bexfield and Plummer (2003)
Vietnam	Red River delta	0.5-510 µg/L	10 µg/L (WHO)	Stopelli *et al*., (2020)

The literature review revealed no study was carried out on groundwater arsenic from the study area. Thus, this is the identified gap in the subject domain. This study aimed to assess the groundwater arsenic concentration from the study area from those identified water sources that are used by local inhabitants for ingestion purposes. Furthermore, to identify the sampling locations which have groundwater arsenic concentration within the acceptable limit of India standard and above it. A comparative analysis with observed arsenic concentration and the percent excess as compared with Indian Standard for drinking water for arsenic. Furthermore, the groundwater arsenic concentration in different aquifer layers. The correlation of groundwater arsenic with other water quality parameters by applying Pearson's correlation coefficient. In addition, the relationship between groundwater, electrical conductivity, and pH. The study outcome will add a new understanding of groundwater arsenic from the study area and its plausible reasons. Moreover, the initiative is to be taken at the local, regional, and, national levels by policy formulation and mechanism for the

implementation of the same and other initiatives to be taken to mitigate arsenic-induced health risks on the local inhabitants of the study area.

Table 2: Groundwater arsenic and the population at risk (Ng *et al*., 2003)

Country or region	Discovery year	Groundwater arsenic concentration (µg/L)	Population at risk (Number)
Taiwan	1950	10-1820	200 000
Chile	1971	900-1040	437 000
Hungary	1974	10-176	220 000
Bangladesh	1980s	<1-4700	50 000 000
India, West Bengal	1980s	<10-3900	1 000 000
Thailand, Ronpibool	1980s	1-5000	1000
China, Xinjiang Province	1980s	1-8000	100 000
Argentina	1981	100-1000	2000 000
Mexico	1983	10-4100	400 000
Peru	1984	500	250 000
USA	1988	10-48 000	Unknown
China, Inner Magnolia	1990s	1-2400	600 000
Bolivia	1997	No data	20 000
Vietnam	2001	1-3050	Millions
Romania	2001	10-176	36000
Nepal	2002	Up to 456	Unknown

MATERIALS AND METHODS

Study area: Chandrapur district (19°25’ N to 20°45’ N and 78°50’ E to 80°10’ E) is situated in the Vidarbha region of Maharashtra state of central India. The district covers an area of 11,364 sq km with elevation ranging from 106 m to 589 m above mean sea level (amsl). The district comprises of 15 administrative blocks. The district is bestowed with natural bounty in the form of dense forest and wildlife on one hand and minerals such as coal, limestone, iron, copper, etc. on other. Due to the abundant presence of natural resources and minerals, the district has witnessed sprawling coal mines, cement industries, pulp and paper industry, and several thermal power plants at the same time Tadoba Andhari Tiger Reserve (TATR) which has one of the largest numbers of tigers in central India (CGWB, 2009).

The climate of the district is characterised by wide climatic conditions ranging from hot summer (May, temperature up to 46 °C) to cold winter (December, temperature up to 7 °C) and general dryness throughout the year. The climate of the district can be classified as a tropical hot. The humidity was observed as 70% during monsoon and 20% in summer. The rainy season had reported rainfall from southwest monsoon (June-September) with annual rainfall ranging from 1200-1450 mm with annual number of rainy days as 60 to 65. The rainfall is asymmetrically distributed in the district. The Worora administrative block receives comparatively

minimum rainfall which gradually increases and reaches a maximum around Bramhapuri administrative block (CGWB, 2009). Geologically, Chandrapur district forms a part of the Gondwana sedimentary basin. Lithologically Chandrapur district presents a variety of stratigraphic units right from Archean to recent alluvium and laterites.

According to the Census of India 2011, the demography of the district comprises 21,94,262 inhabitants out of which 10,73,946 were female and 11,20,316 were male. The decadal growth rate (2001-2011) was found to be 6.0% and the density of the population was 192 people per sq km. The share of the urban population to the total population was found to be 35.1% (Census of India, 2011). The further analysis of Census data sheds light upon the main source of drinking water from the rural area of the Chandrapur district as 36% of inhabitants depend upon hand pumps followed by 7.2% on tube-well which combined is 43.2%. This statistical data highlights, inhabitants from the study area depend mainly upon groundwater as a main source of drinking purpose.

For groundwater sampling site selection criteria from the study area emphasis was laid upon the rural area which largely depends upon hand pump and/or dug well as a major source of drinking, cooking, or other domestic purposes. Further, groundwater sample was collected from different administrative blocks of the district which covers wide geological formations, rainfall, and elevations to understand the distribution of groundwater arsenic. The groundwater sampling was carried out in the post-monsoon season (October).

A total of 36 groundwater sampling locations comprising hand pumps and dug wells from the Chandrapur district were identified for this study which is depicted in Figure 1. Purposive random and stratified sampling was carried out for groundwater sampling from the study area. Of these sampling locations, 34 (94.44%) were from hand pumps and two (5.55%) from dug wells. The grab sampling methodology was adopted to collect groundwater samples.

Groundwater sample for general parameters was collected up to the rim of a narrow mouth polyethylene container of 1000 mL capacity (Poly lab, India) to have no head space into it for entry of atmospheric gases and thus altering its physicochemical properties. The sampling containers' mouth was closed with a screw cap which was afterward sealed with the help of adhesive tape to avoid entry of contaminants into it. The details regarding the sampling location and date were recorded on the sampling container and in the field diary also.

Groundwater temperature alters soon after it gets exposed to the atmospheric environment. Thus, its analysis in the field gives accurate information. Its measurement was carried out in the field itself by using a mercury thermometer with 0.5 ^{o}C division (Gera, GTI, India). Immediately after bringing the groundwater samples into the laboratory they were analysed for different physicochemical parameters as mentioned in Table 3 except for the field analysis parameter. All reagents used while carrying out physicochemical analysis were of AR grade (Merck) and glassware was of borosilicate make. Double distilled water was used for the preparation of reagents. All reagents were prepared as mentioned in APHA (2017). A standardization procedure was carried out and these standardized reagents were used in analysis.

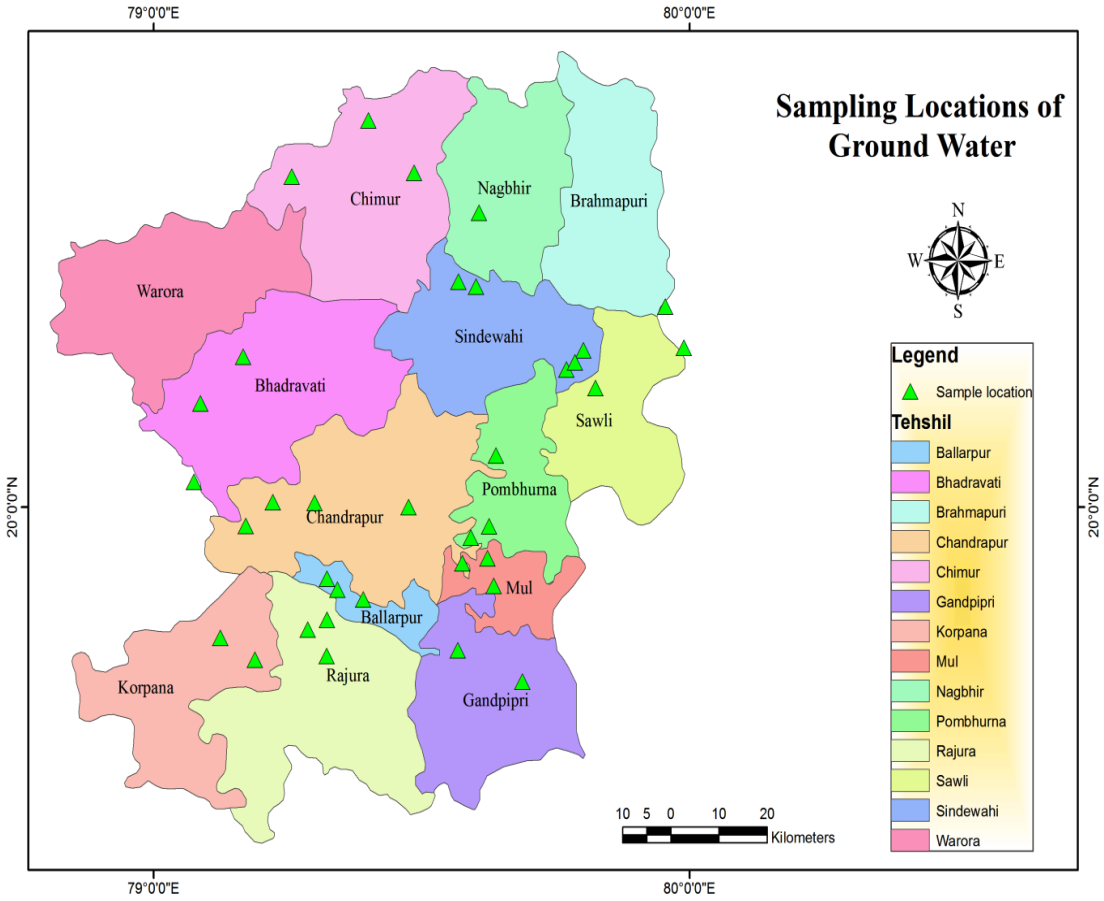

Figure 1: Groundwater sampling locations

For the preservation of heavy metal samples, the addition of concentrated HNO_3 (1 mL for 100 mL groundwater sample, 16 *N*, Merck) into another narrow-mouth polyethylene container (Poly lab, India) was carried out in the field itself. The sampling bottle's mouth was closed with the help of a screw cap which was afterward sealed with adhesive tape to avoid entry of contaminants into it. Before transportation of groundwater samples to the laboratory they were kept in "wet" ice (frozen ice) with double bagged to prevent groundwater damage from melting ice. The groundwater samples were brought to the laboratory for further analysis.

The concentrations of total heavy metals (in this case iron, manganese, and arsenic) from the groundwater samples were determined after acid digestion with concentrated nitric acid (HNO_3). Groundwater samples especially collected for determination of iron, manganese, and arsenic concentrations were acid digested in a pre-leached glass beaker on a hot plate at 95 °C and evaporated to 5 mL without boiling. While carrying out this, glass beakers were covered with a clean watch glass. This process resulted in the total extraction of metals from groundwater. After cooling, into the digested sample a small quantity of 1:1 concentrated HNO_3 (16 *N*, Merck) was added and further refluxed for 15 min to dissolve any precipitate and residue resulting from evaporation. This digested sample after cooling was transferred into 25 mL volumetric flask and diluted up to 25 mL with double distilled water. This acid-digested sample was used for the determination of iron, manganese, and arsenic concentrations from groundwater. Heavy metals analysis was carried out by using ICP-OES (ICP-OES, Perkin Elmer, Germany, Dv 7000) (APHA, 2017; NEERI, 1988).

Table 3: Standard methods used for analysis of groundwater

Parameter	Standard method	APHA (2017), Reference No.	Instrument particular
Colour	Visual Comparison method	B of 2120	NA
Temperature	Mercury thermometer	B of 2550	Gera, GTI, India
pH	Electrometric method	B of 4500-H^+	Digital pH meter, Electronics India, Model 101
Conductivity	Conductivity meter	B of 2510	Digital conductivity meter, Electronics India, Model 601
Total dissolved solids	Total dissolved solids dried at 180 ^oC	C of 2540	Hot air oven, Navyug, India
Alkalinity	Titration method	B of 2320	NA
Total hardness	EDTA titration method	C of 2340	NA
Chloride	Argentometric method	B of 4500-Cl^-	NA
Fluoride	SPANDS method	D of 4500-F^-	Double beam UV/Visible spectrophotometer, Electronics India, Model 1372
Sulphate	Turbidimetric method	E of 4500-SO_4^{2-}	Double beam UV/Visible spectrophotometer, Electronics India, Model 1372
Phosphate	Stannous Chloride method	D of 4500-P	Double beam UV/Visible spectrophotometer, Electronics India, Model 1372
Iron	Inductively Coupled Plasma-OES	C of 3500-Fe	ICP-OES, Perkin Elmer, Germany, Dv 7000
Manganese	Inductively Coupled Plasma-OES	C of 3500-Mn	ICP-OES, Perkin Elmer, Germany, Dv 7000
Arsenic (Total)	Inductively Coupled Plasma-OES	C of 3500-As	ICP-OES, Perkin Elmer, Germany, Dv 7000

NA - Not Applicable.

Table 4: Groundwater quality

Sampling location (Water source)	Temp	pH	EC	TDS	Cl^-	T-Alkal	TH	CH	MH	F^-	Fe	Mn	As
Sonegaon (HP)	30.0	6.87	920	580	20.41	376	136	100	36	1.05	0.136	0.010	0.0305
Telwasa (HP)	30.0	6.85	1210	770	45.94	348	276	272	4	1.10	0.499	0.006	0.0268
Belora (HP)	30.0	7.23	960	600	34.03	376	132	124	8	2.00	0.156	0.032	0.0263
Sagra (DW)	28.0	7.20	1690	1080	108.9	340	340	324	16	0.80	0.080	0.007	0.0312
Pethbhansouli (HP)	28.5	6.96	1290	820	93.58	380	344	240	104	1.05	0.644	0.125	0.0339
Bhisi (HP)	30.0	6.72	1720	1110	137.82	328	540	424	116	1.30	0.698	0.761	0.0251
Pimpalgaon (HP)	30.0	7.18	2830	1820	272.25	388	644	416	228	0.90	1.465	0.008	0.0291
Mowada (HP)	29.5	6.95	1500	960	105.49	340	364	264	100	1.10	0.163	0.003	0.0230
Dongargaon (HP)	30.0	6.83	2420	1560	234.82	332	472	392	80	2.16	0.458	0.125	0.0345
Lohara (HP)	29.0	5.74	330	200	15.31	108	100	84	16	0.50	0.265	0.005	0.0339
Chichpalli (HP)	28.5	6.90	4710	3060	678.93	476	1448	548	900	1.50	0.167	0.087	0.0312
Dabgaon (T.) (HP)	29.5	6.80	2270	1470	209.29	540	548	340	208	1.10	1.627	0.133	0.0314
Naleshwar (HP)	31.5	6.45	1430	910	219.5	228	268	260	8	0.97	0.651	0.009	0.0414
Karwan (HP)	30.0	7.23	960	610	51.04	332	212	96	116	1.76	0.185	0.034	0.0302
Chikmara (HP)	30.5	7.02	1730	1110	129.32	392	552	304	248	1.30	0.084	0.017	0.0334
Pathri (HP)	30.5	6.61	880	560	71.46	232	172	160	12	0.63	0.323	0.035	0.0317
Gunjewahi (DW)	27.5	7.28	460	280	8.50	236	120	52	68	0.74	0.055	0.002	0.0352
Mangali Chak (HP)	29.5	6.83	860	540	28.92	320	220	212	8	0.74	0.144	0.004	0.0334
Govindpur (HP)	30.0	6.89	2390	1540	287.57	416	680	268	412	1.37	0.215	0.005	0.0295
Ratnapur (HP)	31.0	6.86	1510	970	131.02	372	400	340	60	0.78	1.695	0.116	0.0337
Antargaon (HP)	30.5	7.42	1040	670	15.31	440	60	32	28	2.32	0.098	0.003	0.0290

Visapur (HP)	29.5	6.11	770	490	56.15	176	160	100	60	0.93	4.022	0.013	0.0366
Ballarpur (HP)	29.5	6.02	900	570	57.85	160	152	120	32	0.63	3.714	0.026	0.0332
Sasti (HP)	29.5	6.75	3930	2590	353.93	384	1360	852	508	1.10	0.202	0.005	0.0295
Gowari (HP)	30.0	6.93	1700	1100	117.41	408	284	192	92	1.30	0.146	0.002	0.0290
Arvi (HP)	30.5	6.78	1420	920	74.87	280	380	364	16	1.15	0.354	0.006	0.0158
Awarpur (HP)	30.0	7.05	2330	1520	148.03	500	276	184	92	1.82	0.120	0.018	0.0265
Lakhmapur (HP)	30.0	6.92	880	570	13.61	292	284	272	12	1.30	0.124	0.002	0.0308
Kem (T.) (HP)	30.5	7.01	770	490	13.61	280	172	112	60	0.65	1.276	0.022	0.0335
Ganpur (HP)	30.0	6.92	4550	3000	473.04	412	1104	684	456	1.10	0.157	0.002	0.0330
Gondpipari (HP)	31.0	6.85	2300	1500	200.78	408	560	448	112	1.15	0.186	0.082	0.0336
Pombhurna (HP)	30.5	7.04	1850	1200	149.74	432	556	300	256	1.10	0.160	0.004	0.0358
Jam Tukum (HP)	30.0	6.89	2260	1480	215.98	436	344	268	76	1.30	0.115	0.016	0.0365
Dongar Haldi (HP)	30.0	6.95	2700	1770	270.55	580	300	216	84	1.44	0.290	0.044	0.0396
Durgapur (HP)	30.0	6.94	2923	1948	185.13	636	300	272	28	1.48	0.089	0.312	0.0355
Morwa (HP)	29.0	7.03	2010	1310	134.42	332	360	240	120	0.93	0.215	0.003	0.0367

Temp - Temperature in ^{o}C, EC - Electrical conductivity in mmhos/cm, TDS - Total dissolved solids, Cl^{-} - Chloride, T-Alkal - Total alkalinity, TH - Total hardness, CH - Calcium hardness, MH - Magnesium hardness, F^{-} - Fluoride, Fe - Iron, Mn - Manganese, As - Arsenic. All parameters are expressed in mg/L except temperature, pH and EC.

Table 5: Summary of groundwater quality

Parameters	n	Min.	Max.	Average	SD	Variance	Skewness	Kurtosis	Cumulative percentiles				
									25^{th}	50^{th}	75^{th}	95^{th}	98^{th}
Temperature, ^{o}C	36	27.5	31.5	29.8	0.81	0.65	-0.87	1.51	29.5	30	30.1	31	31.2
pH	36	5.74	7.42	6.86	0.33	0.11	-1.64	3.64	6.82	6.91	7.02	7.24	7.32
EC, mmhos/cm	36	330	4710	1788.97	1052.37	1107501	1.21	1.44	950	1600	2307.5	4085	4598
TDS, mg/L	36	200	3060	1157.72	695.84	484201.6	1.21	1.41	595	1025	1505	2692.5	3018
Chloride, mg/L	36	8.5	678.93	149.01	140.91	19857.95	1.91	4.93	49.76	123.36	210.96	383.70	534.80
Total Alkalinity, mg/L as $CaCO_3$	36	108	636	361.55	110.98	12316.60	0.08	0.65	313	374	413	550	596.8
Total Hardness, mg/L as $CaCO_3$	36	60	1448	406.11	320.55	102755.2	1.99	4.15	202	320	542	1168	1386.4
Calcium Hardness, mg/L as $CaCO_3$	36	32	852	274.33	171.66	29467.77	1.40	2.95	151	266	340	582	734.4
Magnesium Hardness, mg/L as $CaCO_3$	36	4	900	132.78	182.42	33278.92	2.70	8.56	25	78	117	469	625.6
Fluoride, mg/L	36	0.5	2.32	1.18	0.42	0.18	0.90	0.74	0.92	1.1	1.31	2.04	2.20
Iron, mg/L	36	0.055	4.022	0.582	0.92	0.84	2.82	8.07	0.14	0.19	0.53	2.19	3.80
Manganese, mg/L	36	0.002	0.761	0.058	0.13	0.018	4.42	21.92	0.004	0.011	0.03	0.177	0.44
Arsenic, mg/L	36	0.015	0.041	0.031	0.004	$2.29e^{-}5$	-0.9385	2.49	0.029	0.032	0.034	0.037	0.040

n - Number of sampling locations, Min. - Minimum, Max. - Maximum, SD - Standard deviation (±), EC - Electrical conductivity, TDS - Total dissolved solids.

RESULTS AND DISCUSSION

The results of groundwater quality obtained from the groundwater samples analysis from different sampling locations from the study area are presented in Table 4. The groundwater quality was assessed based on 10 general parameters and three heavy metals (iron, manganese, and arsenic). The groundwater quality summary of minimum, maximum, average, standard deviation, variance, skewness, kurtosis, and cumulative percentiles is presented in Table 5.

Groundwater arsenic concentration obtained in the study area is divided into three categories (< 0.01 mg/L, 0.01-0.03 mg/L, and 0.03-0.05 mg/L) (Table 6). It is observed that in category I (< 0.01 mg/L), no sampling location fall under this category. In the case of category II (0.01-0.03 mg/L), 13 (36.11%) sampling locations fall under this category. In category III (0.03-0.05 mg/L), a maximum 23 (63.88%) sampling locations had groundwater arsenic concentration. From the table it can be arrived at no sampling location from the study area had groundwater arsenic concentration within the acceptable limit of Indian Standard (0.01 mg/L). The observed groundwater arsenic concentrations over the acceptable limit of the Indian Standard for arsenic (0.01 mg/L) in percent is presented in Table 7. It is observed that the maximum excess percent was in Naleshwar (HP) (0.0314 mg/L, 310%) followed by Dongar Haldi (HP) (0.0296 mg/L, 290%). Minimum excess concentration was observed in Arvi (HP) (0.0058 mg/L, 50%).

Table 6: Groundwater arsenic distribution in the study area

Category I (Less than 0.01 mg/L)	Category II (0.01-0.03 mg/L)	Category III (0.03-0.05 mg/L)
Nil	Sonegaon (0.030), Telwasa (0.026), Belora (0.026), Bhise (0.025), Pimpalgaon (0.029), Mohabala (0.022), Govindpur (0.029), Antargaon (0.028), Sasti (0.029), Gowari (0.028), Arvi (0.015), Awarpur (0.026), Lakhmapur (0.030) (Total : 13, 36.11%)	Sagra (0.031), Pethbhansouli (0.033), Dongargaon (0.034), Lohara (0.033), Chichpalli (0.031), Dabgaon Makta (0.031), Naleshwar (0.041), Karwan (0.030), Chickmara (0.033), Pathari (0.031), Gunjewahi (0.035), Mangali Chak (0.033), Ratnapur (0.033), Visapur (0.036), Ballarpur (0.033), Kem Tukum (0.033), Ganpur (0.032), Gondpipari (0.033), Pombhurna (0.035), Jam Tukum (0.036), Donga Haldi (0.039), Durgapur (0.035), Morwa (0.036) (Total: 23, 63.88%)

The statistical précis of groundwater arsenic from to study area is presented in Table 8. It indicates groundwater arsenic was present in all samples with maximum concentration at Naleshwar (0.041 mg/L, HP) and minimum at Arvi (0.015 mg/L, HP). No sampling location had groundwater arsenic concentration within the acceptable limit of Indian Standard (0.01 mg/L). However, all sampling locations had groundwater arsenic concentration above the acceptable limit. The average groundwater arsenic concentration from the study area was 0.031 mg/L with a standard deviation of ±0.004.

Table 7: Groundwater arsenic vis-à-vis Indian standard

Sampling location (Water source)	Groundwater arsenic concentration, mg/L	Excess (mg/L)	% Excess
Sonegaon (HP)	0.0305	0.0205	205
Telwasa (HP)	0.0268	0.0168	160
Belora (HP)	0.0263	0.0163	160
Sagra (DW)	0.0312	0.0212	210
Pethbhansouli (HP)	0.0339	0.0239	230
Bhisi (HP)	0.0251	0.0151	150
Pimpalgaon (HP)	0.0291	0.0191	190
Mowada (HP)	0.0230	0.0130	120
Dongargaon (HP)	0.0345	0.0245	240
Lohara (HP)	0.0339	0.0239	230
Chichpalli (HP)	0.0312	0.0212	210
Dabgaon (T.) (HP)	0.0314	0.0214	210
Naleshwar (HP)	0.0414	0.0314	310
Karwan (HP)	0.0302	0.0202	200
Chikmara (HP)	0.0334	0.0234	230
Pathri (HP)	0.0317	0.0217	210
Gunjewahi (DW)	0.0352	0.0252	250
Mangali Chak (HP)	0.0334	0.0234	230
Govindpur (HP)	0.0295	0.0195	190
Ratnapur (HP)	0.0337	0.0237	230
Antargaon (HP)	0.0290	0.0190	180
Visapur (HP)	0.0366	0.0266	260
Ballarpur (HP)	0.0332	0.0232	230
Sasti (HP)	0.0295	0.0195	190
Gowari (HP)	0.0290	0.0190	180
Arvi (HP)	0.0158	0.0058	50
Awarpur (HP)	0.0265	0.0165	160
Lakhmapur (HP)	0.0308	0.0208	200
Kem (T.) (HP)	0.0335	0.0235	230
Ganpur (HP)	0.0330	0.0230	220
Gondpipari (HP)	0.0336	0.0236	230
Pombhurna (HP)	0.0358	0.0258	250
Jam Tukum (HP)	0.0365	0.0265	260
Dongar Haldi (HP)	0.0396	0.0296	290
Durgapur (HP)	0.0355	0.0255	250
Morwa (HP)	0.0367	0.0267	260

HP - Hand Pump, DW - Dug Well. (Acceptable limit of Indian Standard 0.01 mg/L)

Table 8: Statistical précis of groundwater arsenic from the study area

Particulars	Details
Total groundwater samples analyzed	36
Groundwater sample from dug well	02 (5.55 %)
Groundwater sample from the hand pump	34 (94.44 %)
Altitude (m amsl) Mean (Range)	211 m amsl (152-287 m amsl)
Groundwater temperature (^oC) Mean (Range)	29.8 ^oC (27.5-31.5 ^oC)
Groundwater samples in which arsenic concentration is detected	36 (100 %)
Groundwater samples in which arsenic concentration is absent	Nil (0.00 %)
Minimum groundwater arsenic concentration	0.015 mg/L (Arvi, HP)
Maximum groundwater arsenic concentration	0.041 mg/L (Naleshwar, HP)
Average groundwater arsenic concentration from the study area	0.031 mg/L
Groundwater samples having an arsenic concentration within the acceptable limit of Indian standard (< 0.01 mg/L)	Nil (0.00 %)
Groundwater samples having arsenic concentration above the acceptable limit of Indian standard (< 0.01 mg/L)	36 (100 %)
Groundwater samples having an arsenic concentration within the permissible limit of Indian standard (< 0.05 mg/L)	36 (100 %)
Arsenic detected in hand pump water samples	34 (100 %)
Arsenic detected in dug well water samples	02 (100 %)
Groundwater arsenic concentration < 0.01 mg/L	Nil (0.00 %)
Groundwater arsenic concentration between 0.01-0.03 mg/L	13 (36.11 %)
Groundwater arsenic concentration between 0.03-0.05 mg/L	23 (63.88 %)
Range	0.015-0.041 mg/L
Standard deviation	±0.004
Kurtosis	2.49

Groundwater arsenic concentration in different aquifer layers (dug well and hand pump) is presented in Table 9. From the table, it is observed that in the dug well maximum groundwater arsenic concentration (0.035 mg/L, Gunjewahi) was comparatively lower than the hand pump (0.041 mg/L, Naleshwar). The reason for this finding can be assigned to the depth of the aquifer. The dug well had shallow depths (35 feet, Gunjewahi) as compared with the hand pump (140 feet, Naleshwar). As a difference in depth below ground level the proximity of different ores and minerals varies which perhaps may have resulted in these findings.

The other water quality parameters which influence the groundwater arsenic concentration were carried out by Pearson's correlation coefficient analysis (Table 10). The groundwater arsenic concentration is negatively influenced by pH, calcium hardness, fluoride, and manganese; whereas, chloride and iron positively influence the concentration.

Table 9: Groundwater arsenic in different aquifer layers

Well type	Minimum arsenic concentration, ppm	Excess as compared to Indian standard*, percent	Maximum arsenic concentration, ppm	Excess as compared to Indian standard*, percent
Dug well	Sagra (0.0312)	210	Gunjewahi (0.035)	250
Hand pump	Arvi (0.015)	50	Naleshwar (0.041)	310

*Indian standard for arsenic acceptable limit 0.01 mg/L.

The relationship between groundwater arsenic, electrical conductivity, and pH is presented in Table 11. For establishing this relationship, the groundwater samples were divided based on the electrical conductivity of <1000 mmhos/cm and >1000 mmhos/cm. From the table, a relationship between pH and groundwater arsenic can be established that in acidic conditions maximum groundwater arsenic is obtained at both electrical conductivity conditions (0.0366 mg/L of arsenic, Visapur, HP, electrical conductivity <1000 mmhos/cm and 0.0414 mg/L arsenic, Naleshwar, HP, electrical conductivity >1000 mmhos/cm). However, the opposite trend was observed for minimum groundwater arsenic concentration. In near-neutral and alkaline conditions minimum groundwater arsenic concentration was recorded.

Table 11: Groundwater arsenic, electrical conductivity, and pH relationship

Electrical conductivity < 1000

Particular	Depth (ft, bgl)	Arsenic (mg/L)	EC (mmhos/cm)	pH
Minimum (Belora, HP)	100	0.0263	960	7.23
Maximum (Visapur, HP)	100	0.0366	770	6.11

Electrical conductivity > 1000

Particular	Depth (ft, bgl)	Arsenic (mg/L)	EC (mmhos/cm)	pH
Minimum (Arvi, HP)	100	0.0158	1420	6.78
Maximum (Naleshwar, HP)	140	0.0414	1430	6.45

The cluster analysis of obtained groundwater arsenic concentrations was attempted by using SPSS (version 16). The result is depicted in Figure 2. From the figure, it can be observed that five different clusters of

groundwater arsenic concentrations can be recognised from the study area. Maximum (n = 16) sampling locations form a major cluster followed by n = 12. The minimum (n = 1, Arvi) sampling location had groundwater arsenic concentration (0.0158 mg/L).

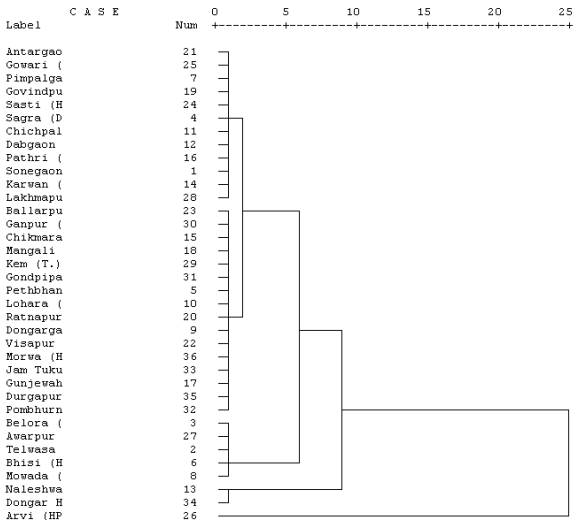

Figure 2: Cluster analysis of groundwater arsenic

The groundwater arsenic concentration with a varying depth of water source below ground level (bgl) is presented in Figure 3. For this analysis, the water source depths were considered. The water sampling depths were divided into six categories of 50 feet depth division. The average of these sampling locations depth and obtained arsenic concentrations were calculated and the obtained values were used. From the figure, it can be seen that at a water source depth of 35 feet bgl arsenic concentration was 0.0339 mg/L which declined further at 92 feet depth. However, the arsenic concentration increased at an average depth of 140 feet (0.0326 mg/L) which further declined at 190 feet (0.0299 mg/L). At an average depth of 250 feet, the arsenic concentration again increased (0.0328 mg/L) and it got reduced to 0.0314 mg/L at an average depth of 300 feet bgl. Thus, below ground level elevated groundwater arsenic concentrations were observed at 35 feet, 140 feet, and 250 feet with a minimum concentration at 190 feet.

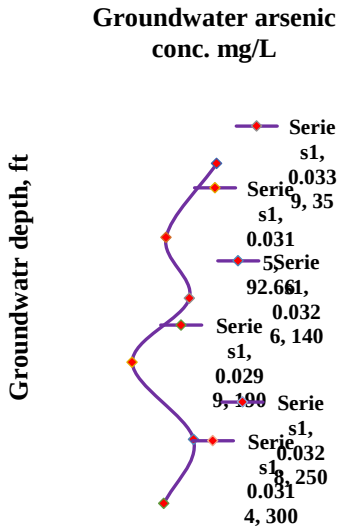

Figure 3: Average groundwater arsenic at varying water source depths

Table 10: Pearson's correlation coefficient

	Temp	pH	EC	TDS	Cl-	T-Alkal	TH	CH	MH	F-	Fe	Mn	As
Temp	1												
pH	-0.0415	1											
EC	-0.0151	0.18885	1										
TDS	-0.0123	0.18686	0.99984	1									
Cl-	-0.0428	0.05778	0.94147	0.93769	1								
T-Alkal	0.11602	0.56933	0.61605	0.61875	0.46272	1							
TH	-0.0863	0.0851	0.88044	0.87718	0.88788	0.34462	1						
CH	0.04763	0.04955	0.81929	0.81882	0.75125	0.29848	0.90477	1					
MH	-0.1954	0.1039	0.79094	0.78579	0.86621	0.32726	0.91808	0.6623	1				
F-	0.20978	0.48259	0.25078	0.25047	0.17145	0.51697	0.07112	0.01106	0.11348	1			
Fe	0.02187	-0.5462	-0.2072	-0.2086	-0.131	-0.3775	-0.1559	-0.1613	-0.1247	-0.3374	1		
Mn	0.04307	-0.0527	0.10996	0.11152	0.07737	0.17936	0.09664	0.18541	-0.007	0.13231	0.04001	1	
As	-0.0236	-0.1979	0.0399	0.04252	0.13025	0.00637	-0.078	-0.1244	-0.0184	-0.218	0.16369	-0.1167	1

The groundwater arsenic concentration in different well structure (shallow well < 100 feet bgl, deep well 101-150 feet bgl, and very deep well 151-300 feet bgl) is presented in Figure 4. From the figure, it can be observed that the maximum groundwater arsenic concentration (0.0326 mg/L) is obtained in a deep well followed by a shallow well (0.0319 mg/L). Very deep well had minimum groundwater arsenic concentration, thus this well-type groundwater is comparatively safe than the other two types of well.

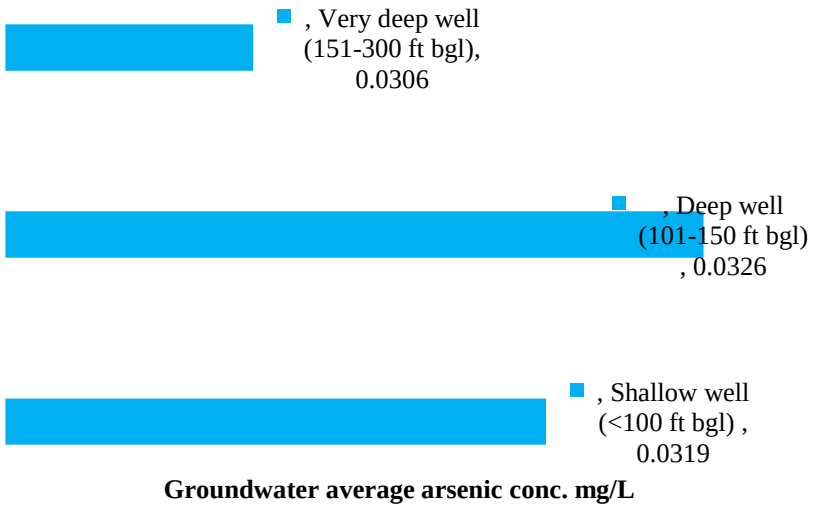

Figure 4: Average groundwater arsenic in different well structure

The predominant form of arsenic is between pH 3 and pH 7 is H_2AsO^{4-}, between pH 7 and pH 11 it is $HAsO_4^{2-}$, and under reducing conditions it is $HAsO_2$(aq) (or H_3AsO_3). Aqueous arsenic in the form of arsenite, arsenate, and organic arsenicals may result from mineral dissolution, industrial discharges, or the application of pesticides. The chemical form of arsenic depends on its source (inorganic, arsenic from minerals, industrial discharges, and pesticides; organic arsenic from industrial discharges, pesticides, and biological action on inorganic arsenic) (APHA, 2017).

The results reported by Zhu *et al*., (2023) state that arsenic concentrations in groundwater in the central Yinchuan basin ranged from 0.7 to 26 µg/L with a mean of 2.19 µg/L, and 5.9% of samples were above 5 µg/L, indicating the arsenic pollution of groundwater. The dissolution of arsenic-bearing minerals in sediment, irrigation water infiltration, and aquifer recharge from the Yellow River were the main sources of arsenic in groundwater.

Rahman *et al*., (2023) reported arsenic concentration from Southwest Bangladesh ranged from 13.10 to 292 µg/L (mean, ±SD : 156.9, ±100.31), which is alarmingly, and 15 times higher than the WHO recommended value for drinking water.

Arsenic concentrations from Huaihe River Plain, China range from 0.001 to 356.00 µg/L, with a median of 2.10 µg/L. The proportion of contaminated shallow groundwater samples is 9.77%, and the counterpart from the deep layer is 2.85%, respectively. Arsenic concentrations are higher in plain areas than those in hilly areas (Xu *et al*., 2022).

The concentration of groundwater arsenic compasses from 0.093 to 0.448 mg/L in pre-monsoon and 0.078 to 0.539 mg/L in post-monsoon; which indicates that all water samples of the Murshidabad district, India exceed the WHO's permissible limit (0.01 mg/L). The particle discharging trends revealed that the Holocene age aquifers are a major contributor of arsenic than Pleistocene age aquifers and this may be the main cause of arsenic vulnerability of both northeast and southwest parts of the study area (Mishra *et al*., 2023).

The concentration of arsenic ranges from 0.46–92.3 µg/L with an average value of 39.4 µg/L, and about 87% of the groundwater samples exceed the given limit of WHO. The Gibbs plot indicates that the significant controlling factor that changes the groundwater chemistry is rock dominance and some of the samples also lie in evaporation crystallization dominance (Rehman *et al*., 2023).

According to Zhou *et al*., (2022) high groundwater arsenic was mostly found in reducing/sub-oxidizing and alkaline environments. Along the south-north groundwater flow path, a significant positive correlation was found between groundwater arsenic, phosphate, and carbonate, suggesting that competitive desorption may have been an important arsenic mobilizing process. Further, a significant negative correlation between groundwater arsenic and the sulphate/chloride ratio and between dissolved organic carbon and the sulphate/chloride was observed.

Goswami *et al*., (2022) reported the seasonal variation in arsenic concentrations with the minimum average concentration in the monsoon season (4.7 µg/L) and the maximum in the post-monsoon season (18.5 µg/L) with 50% of the samples exceeding permissible limits. The differences in the local geological conditions and the groundwater flow may contribute to the spatial variations in mean arsenic concentration in the study area. Results indicate higher arsenic levels associated with a pH range of 6–7 favours arsenic desorption from minerals under reducing conditions.

As conclusions, arsenic is present in all groundwater samples studied from the study area and it was above the acceptable limit of Indian standard and WHO standard (<0.01 mg/L). The elevated groundwater arsenic concentration was mainly distributed in deep wells (hand pumps). The origin of groundwater arsenic can be geogenic in nature. As the study was carried out in post-monsoon season when groundwater level increases after monsoon results in the dilution of the contaminants present in it. Thus, perhaps in summer, elevated groundwater arsenic concentration may be obtained from the sampling locations of the study area.

The hotspots for groundwater arsenic should be identified by local municipal/village authorities from the findings obtained in the study. The water source should be marked with some colour code or suitably so that inhabitants will know that the source is not suitable for drinking purposes. The presence of groundwater arsenic in different aquifers and at varied depths below ground level aid to identify safe aquifer layers and depths below ground level for extraction of groundwater for potable purposes. In groundwater arsenic-dominated areas, an

alternative source of drinking water should be made available by water tankers to reduce the health hazard to the inhabitants.

Awareness among the inhabitants regarding the presence of groundwater arsenic and associated health risks by ingestion of it should be made in the local language (Marathi). A low-cost, environment-friendly, easy-to-understand, and adopt methodology should be developed for the removal of arsenic from the groundwater. Moreover, the methodology should incorporate traditional, advanced, and combined methods. The limitations of the study include, it was carried out in post-monsoon season only and from selected locations. To overcome these limitations a comprehensive study in three seasons (summer, post-monsoon, and winter) and from more sampling locations will provide an in depth understanding of the issue and further will help the local authorities to formulate a policy for future town planning. In addition, regular monitoring of groundwater sources for the presence of arsenic and other contaminants should be carried out by local government authorities.

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